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Test of cross-correlation technique for elastic scattering

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By electronic submission, the Principal Investigator certifies that this information is correct to the best of their knowledge.

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Test of the Cross Correlation Technique for Pulsed Neutron Sources

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Abstract

We propose to continue experiments to investigate the cross correlation technique at pulsed neutron scources, and in particular its efficiency for specialized applications, such a single crystal diffuse scattering with elastic discrimination. Based on our numerical Monte Carlo simulations, this technique can lead to efficiency gains up to two orders of magnitude as compared to traditional techniques using direct time-of-flight spectrometers. However, these simulations do not take into account the influence of backgrounds (from various sources), imperfections in the chopper sequence, or variations of source and/or chopper frequencies. Hence, a thorough experimental test is required to verify the validity of the technique for use in future instrumentation and to provide benchmarks for full experiment simulations. The LANSCE beamline FP12 is ideal for these studies: It has an existing guide with bandwidth chopper as well as a shielded experimental hutch, allowing quick installation of a test experiment. Previous sets of experiments performed in 2009 and 2010 showed that FP12 indeed provides a very low background environment and is perfectly suited for the proposed tests. After modifications to the setup following the first tests in 2009, we were able to successfully reconstruct the full scattering function from a powder sample in 2010. These tests revealed further aspects of the setup that can be improved and that more time is needed to obtain full data sets on single crystal samples. We therefore propose to continue tests of the cross correlation technique with an improved setup and measuring various single crystals with different types of diffuse scattering.

I. Scientific and Technical Purpose

Many recent phenomena of high interest in materials science are driven, or strongly enhanced by complex disorder and nanoscale self-organization in the form of stripes, checkerboard charge/orbital ordering, dimerization *etc*. Complex short range correlations on the 1-100 nm scale are also of great importance on many topics outside materials science, including the design of high-density computer chips, coding theory, protein folding dynamics and many more. A very powerful tool for probing such complex disorder embedded in crystalline materials is single crystal diffuse scattering. It provides a determination not only of the local distortions around a point defect, which can be studied by several other experimental methods, but also the morphology and length scale of defect-defect correlations. There are however technical difficulties that have prevented this branch of neutron scattering from realizing its full potential. Accurate modeling requires measurements over a large volume of three-dimensional reciprocal space, with sufficient momentum resolution to distinguish diffuse scattering from Bragg scattering, sufficient energy resolution to discriminate diffuse scattering from vibrational scattering, and extremely low instrumental backgrounds. Currently, there is no instrumentation that can fulfill all this requirements simultaneously, limiting studies to systems with simple disorder. We have recently started to investigate to usability of the old concept of cross correlation to obtain efficient measurements of elastic diffuse scattering over large volumes.

While the use of statistical choppers in combination with neutron time-of-flight techniques was intensively investigated in the late sixties and early seventies it was quickly found that this technique is not suitable *in general*, and although it was successfully utilized for some problems, its potential for very high gains in efficiency for specific applications was never realized. Based on theoretical considerations and numerical simulations, we propose that this technique indeed has the potential for very high gains in efficiency of up to two orders of magnitude for single crystal diffuses scattering, and eventually also for other types of specialized problems (for a detailed discussion see S. Rosenkranz and R. Osborn, PRAMANA 71, 705 (2008), attached as appendix). In order to verify this concept, which will enable new science in many different areas, we built a prototype instrument that fits within the FP12 hutch and started to perform a series of calibration and test experiments, as well as experiments on known systems with simple and complex disorder. This proposal is a continuation of previous proposals and experiments performed at FP12 in 2009 and 2010. In the first experiments in 2009, we were able to setup the whole instrument within about 3 weeks and to perform preliminary measurements on powders during a second cycle of 3 weeks. From these two cycles, we learned that a proper setup including proper calibrations takes at least one 2-3 weeks, and that proper test-experiments on powders and single crystal samples will require additional 2-3 weeks of measurements each. One of the problems associated with properly aligning the instrument steams from the requirement of a small beam (6mm wide by 25 mm high, dictated by the size of one element of the correlation chopper) and the fact that for diffraction

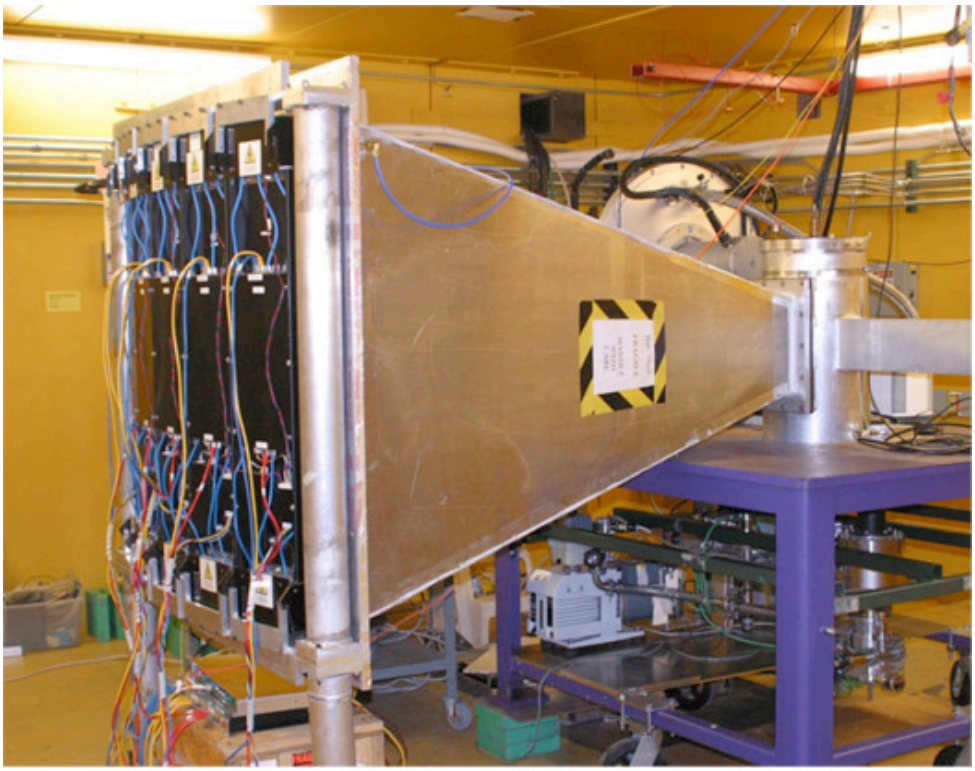

Fig. 1: Previous installation of the prototype inside the FP12 cave. The picture shows the secondary flight path (under He-atmosphere) and the back of the five detector modules, each containing 8 linear position sensitive detectors as well as integrated high voltage, preamp, and detector electronics. The data from the five detector modules is sent in event mode (e.g. each event is registered separately) via Ethernet and Fiber-Optics to the DAS contained in the Rack located outside the cave. Towards the front of the cave, the top of the correlation chopper is visible.

experiments the whole table and in particular the center of sample rotation has to be aligned to the center of the beam to within a fraction of these dimensions. Accurate determinations of the chopper phase-offset and Moderator-to-sample distance furthermore require several measurements (each 12-24h) with the chopper phased to the source at different phases and speeds. In the second sets of experiments performed last year we used an improved setup with which we were able to reconstruct the full scattering function S(|Q|,ω) form a powder sample of $PrAl_3$.This sample has a strong crystal field transitions at 4.5meV energy transfer which we were able to clearly resolve with a measurement with the correlation chopper running at 7393 RPM and an exposure of 105h. It should be noted that with our setup, we have to mask the beam down to an approximately 6mm x 25mm size, from which we loose about a factor 50-100 of the incident flux. An instrument designed for such smaller beams and using focusing guides would therefore be very competitive even for such inelastic experiments, in particular since spectra are obtained over a large range of momentum transfer. More importantly however, this first test clearly demonstrates the ability of the technique to obtain energy discrimination of the elastic signal with high enough resolution to distinguish inelastic processes in the order of a few meV (phonons).

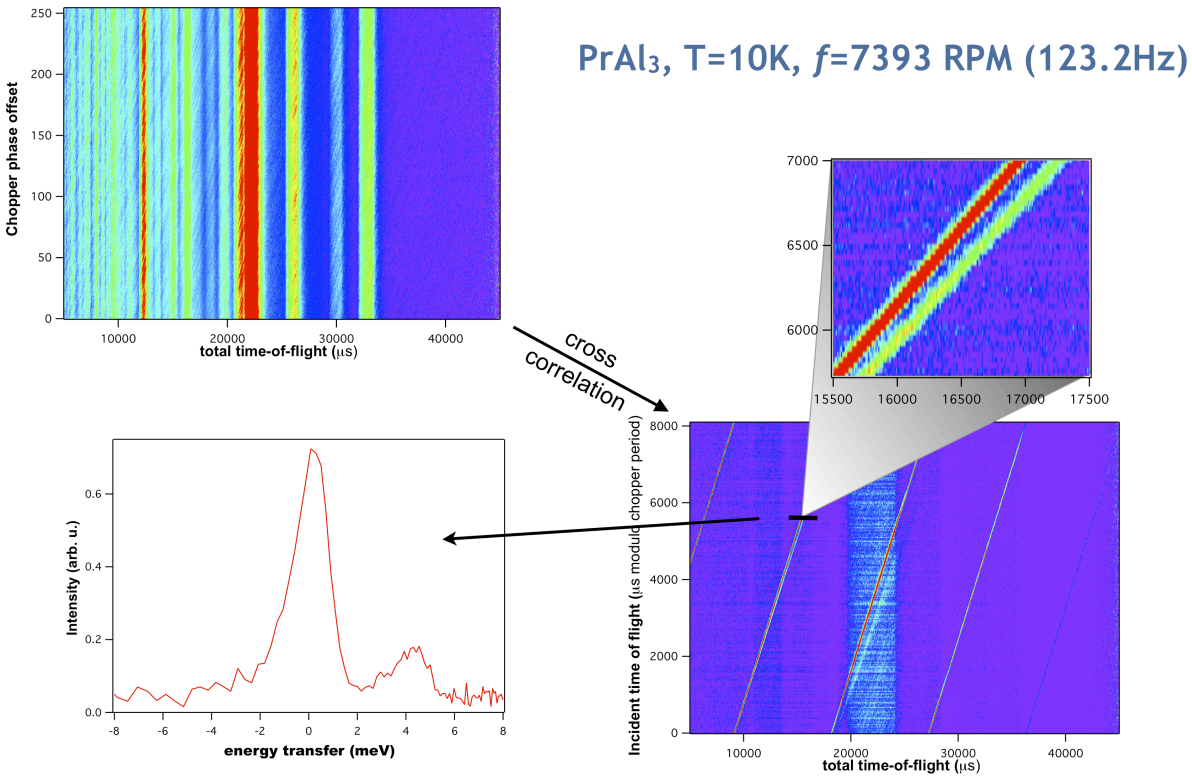

Fig. 2: Raw data (top left) as a function of total time of flight and chopper phase-offset measured for $PrAl_3$ with the chopper running asynchronously to the neutron source at 7393 RPM. The bottom right shows the scattering function reconstructed through cross correlating the raw data with the modulation sequence. The zoomed-in region shown in the panel above clearly reveals the separation of the inelastic transition from the elastic line. The panel at the bottom left shows a cut along the total time-of-flight for fixed incident energy, corresponding to a spectrum as measured on a traditional direct geometry time-of-flight spectrometer. The elastic line as well as the crystal field transition at ~ 4meV are both clearly resolved, demonstrating the ability to obtain energy discrimination in the order of meV.

An important improvement was made to the setup of the secondary flight path. During the first experiments in 2009, we found severe problems with scattering from strong Bragg peaks from an Al-sheet in front of the detectors. This originally 1/8" thick Al-sheet covering the back of the secondary flight path was necessary in order to keep a He-atmosphere to reduce background from air scattering. This sheet was straight and because of the curvature of the detector mount was approximately 4-5" away from the detector in the center of the array. While performing measurements with single crystals (that is to measure the diffuse scattering in the vicinity of a Bragg peak), the bragg peak itself necessarily has to be brought into reflection condition, and this leads to secondary scattering of the strong Bragg-peak beam from the Al in the form of powder rings originating from this Al-sheet. This scattering from Al then contaminated the whole region where diffuse scattering was expected, and was so strong that it overshadowed any diffuse scattering form the sample. We therefore removed this Al-sheet and constructed a cover of the back of the secondary flight path with aluminized Mylar foil. With this new setup, test experiments with single crystals in Bragg condition did no longer show secondary scattering of the Bragg peak from the Al and we were able to observe diffuse scattering from a Calcium stabilized Zirconia sample. These systems have a very well characterized, strong diffuse scattering. While we have observed the expected signatures of this diffuses scattering, detailed analysis of the data for comparison with previous experiments and theoretical studies is in progress. This comparison should reveal how well the correlation technique works for systems that like Zirconia have very strong, broad diffuse scattering features that are well separated from the Bragg peaks. We were however not able to perform measurements to test weak diffuse scattering, such as magnetic diffuse scattering in frustrated magnets, or diffuses scattering close to Bragg peaks, such as Huang scattering from locale defects such as trapped polarons, due to problems with the cooling for the beamline bandwidth chopper and because of the long measurement time required (hundreds of hours per setting for a weak signal). We therefore plan to use the same setup, with further minor improvements, to perform conclusive tests using single crystal samples with different types of diffuse scattering.

II. Timeline

Following our experience with the previous tests performed on FP12 we request 60 days of beamtime in the fall of this year in order to setup and test the equipment, and in order to perform the necessary calibration and test experiments. The setup will be the same as previously used with minor improvements in the setup.

Week 1-3: Installation, calibration, and commissioning

- Installation and positioning using neutron camera of incident beam slits and shielding
- Installation and accurate optical positioning of sample table
- Installation and positioning of chopper
- Installation of sample vacuum chamber
- Installation of beam monitors and hookup to DAS electronics
- Accurate positioning of sample chamber using neutron camera and beam monitors
- Installation of secondary flight path
- Installation of detector modules, DAS electronics
- Hookup of detectors, communications electronics, DAS and chopper

- Power-up and test of DAS
- Installation and testing of shielding of anything in incident beam and detector backside:
  Note that in last setup, we found that most of the background comes from thermal neutrons reflecting off from the floor, walls and roof of the hutch (which is steel), which is why shielding around detector and secondary flight path has to be carefully placed and tested. This is crucial to reduce background low enough in order to be able to observe weak diffuse scattering, even though the secondary flight path as well as the detector modules have an inernal ¼" B4C crispy-mix shielding already incorporated.
- Power-up and testing and calibration of phase offset of beamline frame-overlap choppers. We found in the previous tests that the phase-offset calibration is frequency dependent and therefore have to perform runs with 5-6 phase offsets at several frequencies, requiring a minium of about 6-12h each.
- Diffraction-commissioning of DAS through runs with Vanadium-sample.
- Commissioning and calibration of DAS using powder standards (e.g. Si) for diffraction.
- Normalization runs with Vanadium, chopper running asynchronously
- Background run of empty instrument, chopper running asynchronously

Weeks 4-9: Assessment of the cross correlation technique at pulsed sources utilizing single crystal samples. Last years test successfully provided first measurements on powder samples that we will further use for benchmarking the accuracy of our numerical simulations during this summer. The most important test are however measurements planned on single crystals to test the performance of different types of diffuse scattering, representing different resolution requirements, different ratios of coherent to incoherent scattering, degree of inelasticity *etc*, and are required to obtain a realistic assessment of the performance of the technique.

- Test performance of the cross correlation technique for measuring strong diffuse scattering from defect structure utilizing Calcium stabilized Zirconia, e.g. ZrO_2 (18 mol% Ca_2O_3). This system is ideal for these tests for several reasons:
  - Because of the large defect concentration, the diffuse scattering is very strong
  - single crystals are available and have already been measured previously
  - These are some of the most intensely studied systems (with regards to the diffuse scattering), and hence provide a good benchmark

  First measurements from Zirconia samples at room temperature were performed in our last experiments in December of 2010. A detailed analysis of the RT data currently in progress will reveal whether further room-temperature measurements are necessary. We also plan additional measurements at low temperature for comparison of how well the TDS suppression utilizing the cross correlation method works.
- Test performance of the cross correlation technique for measuring he strain-field induced by local distortions (Huang scattering), e.g. in the bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$. This type of scattering is strongly peaked at the Brillouin zone center (e.g. the Bragg peak position), and falls of in intensity as a power-law from the Bragg peaks themselves, with a characteristic shape determined by the symmetry of the unit cell distortion. We have extensively studied this Huang scattering in the bilayer manganites ourselves and have high quality single crystals available. In these samples, the scattering is weak, in the order of the thermal diffuse scattering, which we

wish to discriminate against, and four to five orders of magnitude weaker than the Bragg Peak intensity, hence presenting the most realistic benchmark for the testing the cross correlation technique. The bilayer manganites also exhibit weak, incommensurate, broad correlations due to short-range charge/orbital ordering allowing to asses the performance of the cross correlations technique to measure this type of complex disorder of great importance to many strongly correlated electron systems.

- Test performance of the cross correlation technique for measuring extended short-range correlations utilizing relaxor ferroelectrics, e.g. $PbZn_{1/3}Nb_{2/3}O_3$. These systems are known to form polar domains in the 5-10nm scale and exhibit extended rods of diffuse scattering along certain (*hh*0) directions. These systems are again well studied, high quality single crystals are available, and the diffuse scattering is relatively strong.

- Test performance of the cross correlation technique for measuring broad magnetic diffuse scattering in frustrated systems, e.g. $TbTi_2O_7$. This system has well characterized diffuse scattering at low temperature. A check performed during the 2010 experiments showed that we should have enough counts in the Bragg peak within a few days in order to be able to see the diffuse scattering.

Appendix:

Efficient Single Crystal Diffraction with Elastic Discrimination

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Abstract

Single crystal diffuse scattering provides one of the most powerful probes of short-range correlations on the 1 to 100 nanometer scale, which often are responsible for the extreme field response of many emerging phenomena of great interest. Accurate modeling of such complex disorder from diffuse scattering data however puts stringent experimental demands, requiring measurements over large volumes of reciprocal space with sufficient momentum and energy resolution. Here, we discuss the potential of the cross correlation technique for efficient measurement of single crystal diffuse scattering with energy discrimination, as will be implemented in a novel instrument, Corelli. Utilizing full experiment simulations, we show that this technique readily leads up to a fifty-fold gain in efficiency, as compared to traditional methods, for measuring single crystal diffuse scattering over volumes of reciprocal space with elastic discrimination.

Introduction

Many phenomena of current interest are controlled by the introduction of complex disorder in crystalline solids, characterized by short-range correlations that extend from 1 to 100 nanometers [1]. Examples where disorder, and the associated nanoscale self-organization, results from the competition between groundstates with incompatible order include colossal magnetoresistance, relaxor ferroelectricity, negative thermal expansion, quantum spin liquids, and high temperature superconductivity. Obtaining a detailed understanding of such complex disorder is therefore of great importance in condensed matter, with potential for impact on many other fields [2].

Single crystal diffuse scattering has the potential to provide one of the most powerful probe of complex disorder as it not only measures the existence and morphology of local distortions, but also directly probes defect-defect correlations, i.e., the tendency for defects to cluster into nanoscale ordered

structures [3-6 and references therein]. However, obtaining a realistic physical understanding of how a material is locally organized puts stringent experimental demands, usually requiring measurements over large volumes of reciprocal space with sufficient momentum and energy resolution to distinguish the various scattering constituents. Current instrumentation is either suited to measurements over large momentum range without energy discrimination, such as time-of-flight white-beam Laue instruments, or provides sufficient energy discrimination but only over a limited range of momentum space. In order to overcome these limitations we propose to implement the old idea of using cross correlation to obtain energy discrimination in a dedicated diffuse scattering instrument at a pulsed neutron source to provide efficient measurements of diffuse scattering with both coverage of large volumes of momentum space and sufficiently high energy resolution.

Cross Correlation

The goal of the cross correlation technique is to maximize the utilization of the available neutron flux by exploiting modulation of the incident neutron beam. The use of this technique for neutron scattering was intensively investigated in the late sixties and early seventies, mainly relating to steady state sources and beam modulations obtained using mechanical choppers [7-10], but it was also adapted for periodically modulated sources, such as long pulse reactors available at the time [11-12]. This generalization of the technique can be applied equally well to the new generation of short-pulsed spallation neutron sources and a proof of principle was later carried out in the mid eighties at IPNS [13]. The possibilities and benefits of utilizing modulation of the polarization was also discussed early on [14,15] and used in few experiments.

The cross correlation technique differs from traditional time-of-flight spectrometers, which only use a very narrow band of fixed incident or final energy, in that the incident neutron pulse is modulated in time in a pseudo-random fashion. The signal recorded at the detector is then no longer directly proportional to the sample scattering function, but he latter can be reconstructed by forming the cross correlation of the measured data with the modulation sequence, allowing utilization of up to 50% of the incident wavelength spectrum compared to the typically 1-5% of a traditional time-of-flight spectrometer. But because of the cross-correlation, the statistical errors of the derived scattering function are now correlated among different channels leading to poor statistics for weak signals, which has lead to the demise of the correlation technique for general spectroscopy. At a pulsed source however, the

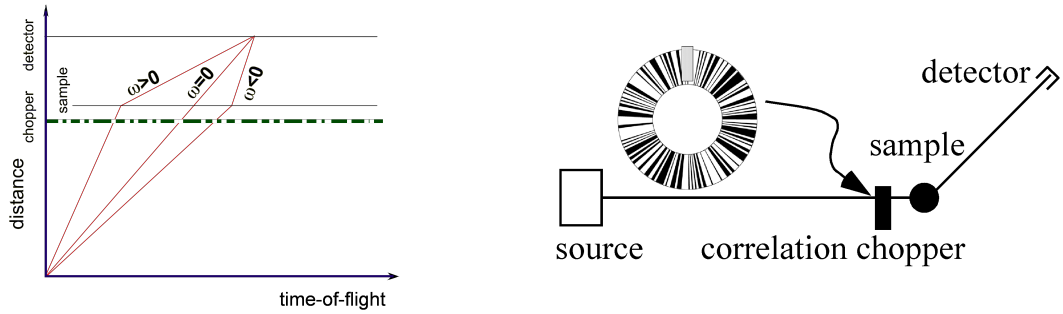

Figure 1: Illustration of the intensity measured with the cross correlation technique at fixed total time-of-flight and schematic of the simplified instrument used for the simulations.

cross correlation is performed only across the incident time-of-flight, meaning that errors in the reconstructed S(Q(ω), ω) are only correlated along the energy transfer ω, but for a fixed energy transfer ω are independent in the momentum transfer **Q**. A strong signal that is localized in momentum space, such as a Bragg peak, therefore only aversely affects the statistics in a small volume around the Bragg peak position. The correlation method therefore has a tremendous potential for providing efficient measurements of single crystal diffuse scattering over large volumes of reciprocal space with elastic discrimination, where we wish to discriminate inelastic scattering, e.g. phonon scattering, that is in the same order of magnitude or weaker than the elastic signal.

Full Experiment Simulations and Discussion

In order to prove the feasibility and obtain a realistic estimate of the gain in efficiency achievable by utilizing the cross correlation to obtain energy discrimination for single crystal diffuse scattering, we performed full experiment simulations utilizing McStas [16] with specialized routines for chopper and sample components. For this purpose we use a simplified instrument, as schematically shown in Figure, that incorporates only the source, a pseudorandom chopper, a sample, and a single point detector at a fixed scattering angle. For the simulations we utilized parameters relevant to Corelli, the dedicated single crystal diffuse scattering instrument under development at SNS, i.e., facing the high-resolution water moderator [17], a disk chopper with a radius of 35 cm running at 350 Hz and a 255 element pseudorandom sequence 19m form the moderator, the sample at 1 m from the chopper, and a 1cm x 1cm detector 2.5 m from the sample. We first simulate a powder sample using a harmonic oscillator model with $\hbar\omega_0$=2meV, including excitations up to the 5th harmonic.

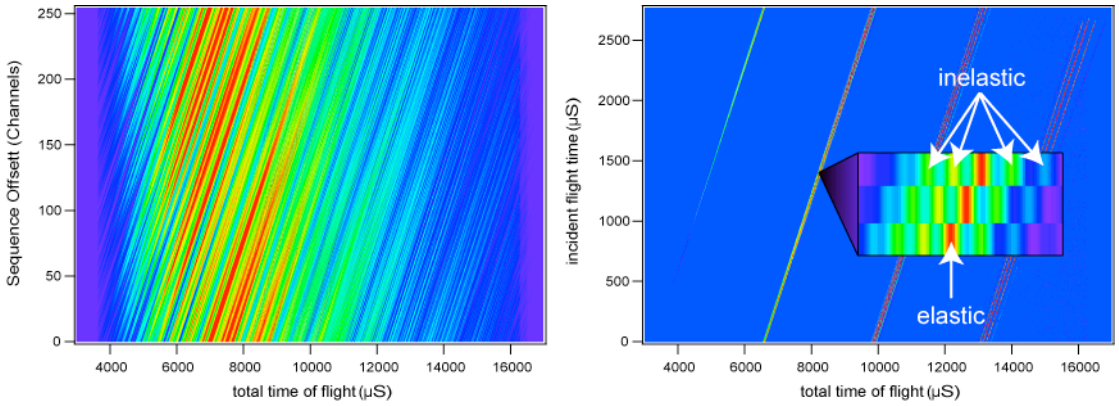

Figure 2: Intensity measured by the detector as function of total time-of-flight and correlation sequence phase offset and the corresponding cross correlated data obtained for the full experiment simulations using a harmonic oscillators sample.

Figure 2 shows the resulting intensity for a single point detector as a function of total time-of-flight and phase-offset of the pseudorandom sequence with respect to the source pulse. Since the cross-correlation is performed over the phase-offset, it is necessary to sample the total time-of-flight spectrum for all possible offsets with the same probability, which is easily and almost instantaneously obtained by running the chopper asynchronously with the source. The right panel of Fig. 2 shows the cross-correlated data, which clearly demonstrates the reconstruction of the scattering function with the elastic scattering clearly separated from the inelastic excitations. This is seen more clearly in Figure 3, which

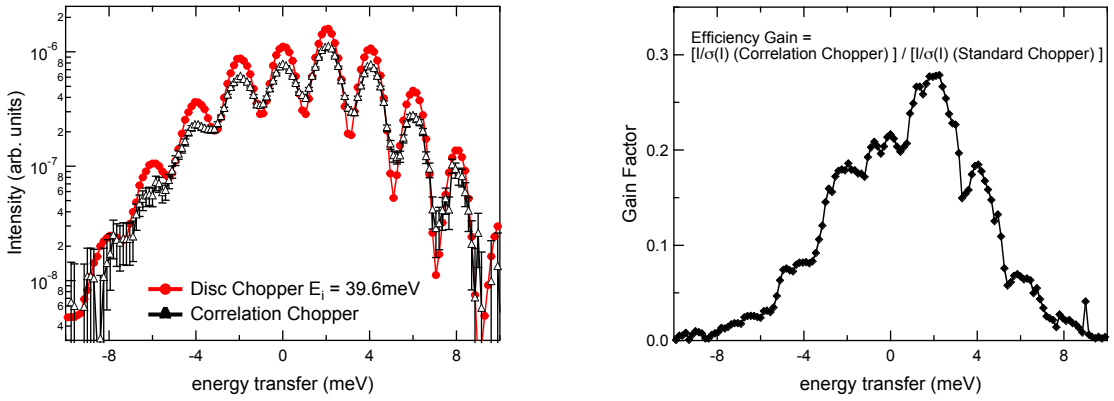

Figure 3: Comparison of a single spectrum with fixed incident energy extracted from the cross correlated data with the spectrum measured with a traditional single-aperture chopper and corresponding gain in efficiency of the cross correlation

compares a single spectrum with constant incident energy extracted from the cross correlated data from the enlarged region shown in Fig. 2, with the spectrum obtain using a conventional, single-aperture chopper with a corresponding fixed incident energy. The conventional chopper is clearly able to resolve all excitations up to and including the 5^{th} harmonic, whereas the correlation chopper surprisingly can still resolve up to the 4^{th} harmonic on the neutron energy loss side, which is more than an order of magnitude weaker than the strongest signal, and the 3^{rd} harmonic on the energy loss side is just still visible. For this comparison of a single spectrum, the cross correlation is always less efficient than the traditional chopper as seen from the gain factor shown in Fig. 2, which is always < 1. The cross correlation technique however simultaneously measures a wide band of incident energies, i.e., provides hundreds of spectra corresponding to varying incident energies at once. This in turn means that the elastic intensity $S(Q,\omega=0)$, which we are interested in, is obtained over a large range of momentum transfer at once, resulting in an actual total gain in efficiency of the cross correlation method of the order of 30-50 for measuring the elastic scattering signal with sufficient energy discrimination.

We further illustrate this gain in efficiency obtained from the simultaneous coverage of a large range of momentum transfer of even a single detector pixel by extracting the elastic intensity form the simulated data. For this purpose, we simulate a perfect incoherent scatterer, for which the total scattering $S(Q)$ is constant as a function of momentum transfer, whereas the elastic intensity decays according to the Debye-Waller factor $S(Q,\omega=0) \sim \exp(-2W(Q))$ with the remaining intensity distributed over the inelastic channels. Here, we use a simplified scattering function with all the inelastic scattering in a single excitation at ±5 meV. We furthermore include a set of Bragg peaks, simulating a cubic sample with lattice constant a = 3.875 Å, aligned such that the (h,0,0) peaks are in Bragg condition at a scattering angle of 90°, with the intensity of the Bragg peaks chosen to be three orders of magnitude stronger then the incoherent scattering at Q = (4,0,0). The cross correlated data obtained from these simulations, Fig. 4, shows streaks along the cross correlation direction at the Bragg peak positions. This is because the color scale of this figure is chosen such that the incoherent scattering remains visible and thus the Bragg peaks are strongly oversaturated. The streaks then appear because of the poor statistics of the cross correlation in the presence of the very strong Bragg peaks. The true signal along these streaks would be zero, except for the incoherent scattering at 0 and ±5meV, but in the cross correlation it is reconstructed as the difference of two large numbers, both in the order of the Bragg peak intensity, whose fluctuations are still stronger than the incoherent signal, giving rise to the streaks. The Bragg peaks however extend only over a very small range and therefore only affect a limited range of the data.

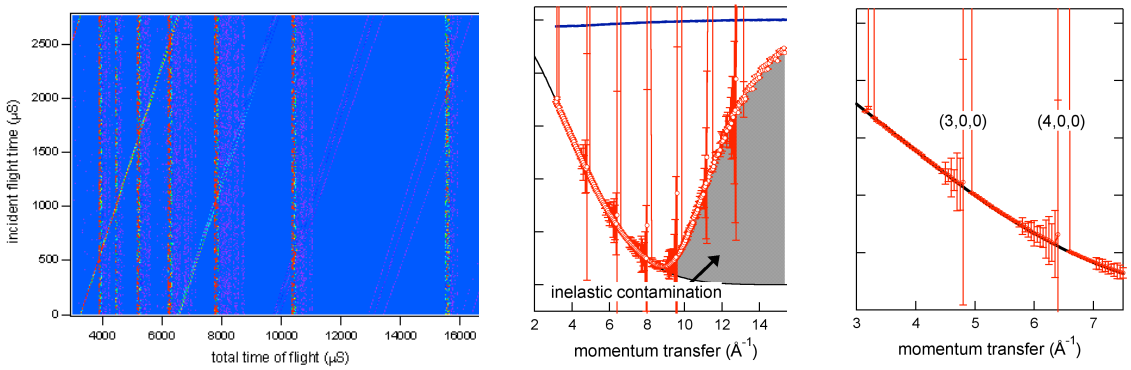

Figure 4: Full experiment simulation using a perfect coherent scatterer and including Bragg peaks. The middle panel shows the elastic scattering extracted from the cross correlated data in red and the total scattering S(Q in blue. The right panel shows an enlarged region from the middle panel around the (3,0,0) and (4,0,0) Bragg peaks.

This is clearly seen in the middle and right panels of Fig. 4, which show the elastic intensity S(Q,ω=0) extracted from the cross-correlated data.

We first note, that the cross correlation successfully provides the correct elastic intensity over a large range of momentum transfer as the extracted S(Q,ω=0), represented by the red circles is identical to the theoretical intensity, shown as a black line, up to Q~9 Å^{-1} for a single oint detector at a scattering angle of 90°. This elastic intensity is very different from the total scattering S(Q), shown in blue, which for this sample is constant and is the quantity measured with a standard, energy integrating diffractometer. At higher momentum transfer, the elastic intensity extracted from the cross correlated data starts to deviate from the Debye Waller curve, approaching the total intensity S(Q). This is because the energy resolution decreases with decreasing total time-of-flight and hence increasing momentum transfer. Upon increasing Q the energy discrimination therefore becomes increasingly less effective and inelastic contamination finally leads to the deviation from the pure elastic signal towards the fully energy integrated S(Q).

Close to the Bragg peak positions, the statistics of the elastic intensity extracted from the cross correlated data becomes very poor. This is due to Bragg peak intensity leaking into the inelastic channels around the Bragg peak position due to finite energy resolution and sample mosaic. The usable range of momentum transfer over which the elastic intensity can be measured at once with the correlation method is however still considerably large even for this worst-case detector position exposed to a series of Bragg peaks. For comparison, a traditional single-aperture chopper instrument would only measure a single Q-point. We further note that the elastic intensity can be extracted without noticeable

decrease in statistics even when it is less than 20% of the total S(Q), which for the example used here is the case for $Q > 6Å^{-1}$. As is seen from the right panel of Figure 4, the extracted elastic intensity shows appreciable statistical errors only when it becomes contaminated from nearby Bragg peaks.

Last but not least, we point out that the examples shown here represent worst-case detector positions, which actually contain Bragg peaks. The poor statistics of the elastic signal because of nearby Bragg peaks is by far the biggest concern for the efficiency of the cross correlation for obtaining energy discrimination in diffuse scattering. In the absence of a Bragg peak, the intensity of the elastic signal, even for weak diffuse scattering, is usually in the order of the thermal diffuse scattering and therefore not small compared to the average signal over the whole energy spectrum, over which the cross correlation is performed. And as we demonstrated above, the statistics of the elastic signal is not strongly degraded even when it only makes up ~ 10% of the total scattering. For single crystals the cross correlation will provide poor statistics only over a very limited range of momentum transfer surrounding the Bragg peaks, the extent of which depends on beam divergences, sample size, mosaic and $\Delta d/d$. For the very large volume of reciprocal space not affected by this, our simulations prove that the cross correlation at pulsed neutron sources provides a very efficient method to obtain energy discrimination in single crystal diffuse scattering.

Acknowledgement

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